The phase transition in the localized ferromagnet EuO probed by μSR

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We report results of muon spin rotation measurements performed on the ferromagnetic semiconductor EuO, which is one of the best approximations to a localized ferromagnet. We argue that implanted muons are sensitive to the internal field primarily through a combination of hyperfine and Lorentz fields. The temperature dependences of the internal field and the relaxation rate have been measured and are compared with previous theoretical predictions.

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Europium oxide (EuO) crystallises in the rock-salt structure and is a ferromagnetic semiconductor with a Curie temperature (T_C) of 69 K [1]. It shows a colossal magnetoresistance effect in its Eu-rich form [2, 3] and the associated metal-insulator transition has been linked to the formation of bound magnetic polarons [4]. It is the only magnetic binary oxide known to be thermodynamically stable in contact with silicon [5], and this, together with a nearly 100% spin polarization of mobile electrons for carrier concentrations below half filling of the conduction band [6, 7], means that it is thought to be highly relevant for spintronic applications [8]. The magnetic moments of the Eu^{2+} ($4f^7$, $^8S_{7/2}$) ions result from 4f charge density which is nearly completely localized inside the filled $5s^25p^6$ shells and there is negligible overlap. This makes EuO an excellent approximation to a Heisenberg ferromagnet [9], although there is evidence of some momentum dependence in the exchange interactions [10]. The ferromagnetic interactions are due to an indirect exchange mediated by electrons in anion valence bands [11], specifically virtual excitations of oxygen valence band p electrons into empty Eu^{2+} (5d) conduction bands and exchange interaction of the d electron (p hole) with the localized 4f electrons. EuO is one of a family of isostructural europium chalcogenides EuX with X=O, S, Se and Te. As the size of the chalcogen increases from O to Te, the lattice parameter a increases across the series: 5.14 Å in EuO, 5.97 Å in EuS, 6.20 Å in EuSe and 6.60 Å in EuTe [12]. Only EuO and EuS are ferromagnets and both the nearest-neighbor and next-nearest-neighbour exchange constants are larger in EuO compared to EuS [9, 13]. In EuO, the saturation magnetization M_s is given by $gJ\mu_B(4/a^3)$, yielding $\mu_0 M_s = 2.40$ T (using $gJ = 7$).

Neutron studies of EuO are significantly hindered by the strong absorption of neutrons by Eu, though the use of a thin-slab geometry and use of ^{153}Eu (which has the smaller absorption cross section of the two naturally occurring isotopes) has allowed a detailed study to be performed [9]. Nevertheless, a technique such as muon-spin rotation (μSR) avoids these difficulties entirely, and in contrast to NMR there is no electric field gradient or quadrupolar contribution to the observed muon response, simplifying the analysis. In this paper we present the results of μSR experiments to study the magnetic order and the fluctuations in samples of EuO and use the results to compare with theoretical predictions [14–16].

Our μSR experiments were carried out using the GPS instrument at the Swiss Muon Source (SμS), Paul Scherrer Institute (PSI) in Switzerland. In our μSR experiment, spin polarised positive muons (μ^+, momentum 28 MeV/c) were implanted into small crystals of EuO. The muons stop quickly (in $< 10^{-9}$ s), without significant loss of spin-polarization. The observed quantity is then the time evolution of the average muon spin polarization $P_z(t)$, which can be detected by counting emitted decay positrons forward (f) and backward (b) of the initial muon spin direction; this is possible due to the asymmetric nature of the muon decay [17, 18], which takes place in a mean time of 2.2 μs. In our experiments positrons are detected by using scintillation counters placed in front of and behind the sample. We record the number of positrons detected by forward (N_f) and backward (N_b) counters as a function of time and calculate the asymmetry function, $G_z(t)$, using $G_z(t) = [N_f(t) - \alpha_{exp} N_b(t)]/[N_f(t) + \alpha_{exp} N_b(t)]$, where α_{exp} is an experimental calibration constant and differs from unity due to non-uniform detector efficiency. The quantity $G_z(t)$ is then proportional to $P_z(t)$.

Raw data for EuO at two temperatures, one slightly higher and one much lower than T_C, are shown in Fig. 1(a). These demonstrate that spin relaxation above T_C changes to a damped coherent oscillation below T_C. If the experiment is repeated with a sample of EuO in which 0.6% of the Eu ions are replaced with Gd, an almost identical result is obtained [Fig. 1(b)] although the damping rate of the oscillations in the ordered state is noticeably larger (by about a factor of two). This level of Gd doping is known to introduce a static magnetic inhomogeneity [19]. The frequency of the oscillations in both cases increases rapidly on cooling below T_C, see Fig. 1(c,d,e), approaching ≈ 30 MHz at zero temperature. The Fast Fourier transform data in Fig. 1(c) demonstrate that only a single precession frequency is observed. The tempera-

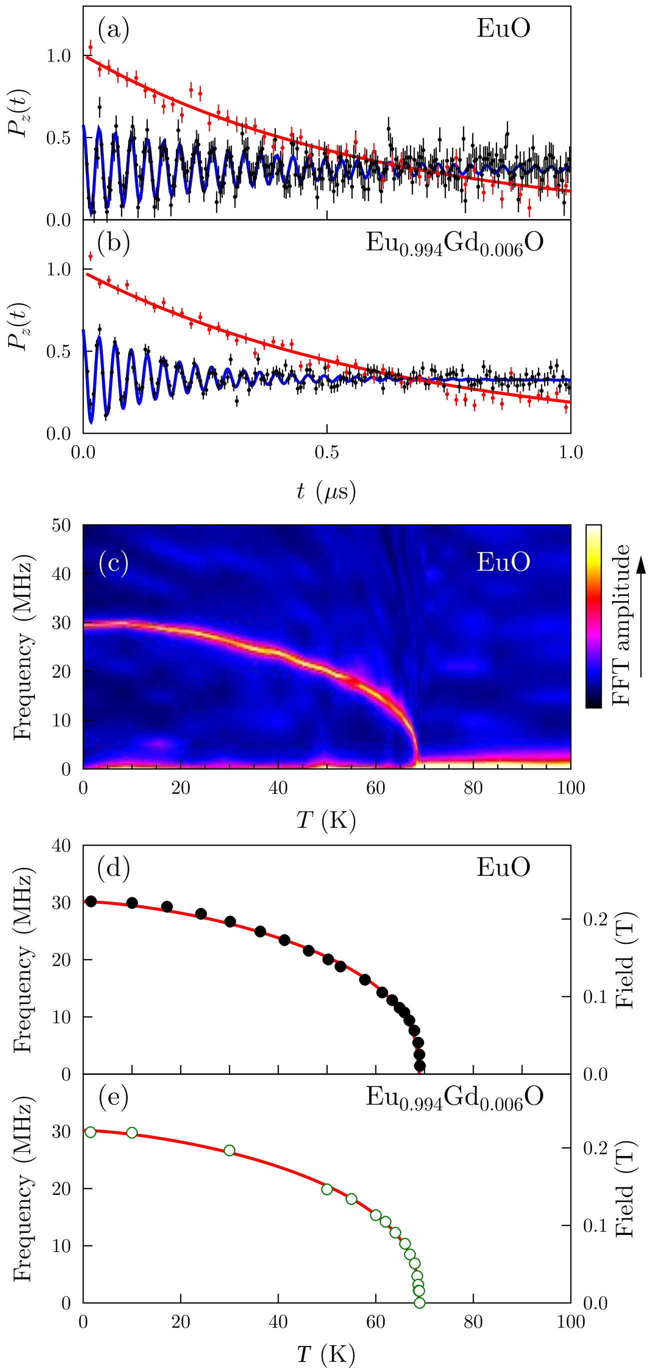

FIG. 1: (Color online.) (a) Raw μSR data for EuO. Above $T_{\rm C}$ (at 70 K), $P_z(t)$ relaxes. An oscillating signal develops below $T_{\rm C}$ (data shown for 1.5 K). (b) The same for $Eu_{0.994}Gd_{0.006}O$. (c) Plot showing FFT of the muon data for EuO as a function of temperature. The single precession frequency which follows the order parameter is clearly visible. (d) The extracted precession frequency for EuO as a function of temperature. (e) The same experiment repeated for $Eu_{0.994}Gd_{0.006}O$. The line through the data in (d) and (e) is identical for the two samples and is a best fit of the data to the phenomelogical function $\nu(T) = \nu(0)(1-(T/T_{\rm C})^{\alpha})^{\beta}$, producing $\alpha \approx 1.5$, $\beta \approx 0.4$). A more reliable extraction of the critical exponent β, focussing only on the critical regime, is given later in the paper.

ture dependence of the precession frequency in the Gd-doped sample is almost identical to that in the pure sample, demonstrating that the order parameter is following the intrinsic magnetism in the EuO host and is relatively insensitive to low levels of doping. In fact, at the 0.6% level of Gd-doping, 93% of the Eu ions have their full complement (twelve) of nearest-neighbour Eu ions, with most of the remaining Eu ions having only one of those nearest neighbours replaced by Gd. For both samples, the relaxation rate of the oscillatory component rises as T approaches $T_{\rm C}$ from below and the low-temperature relaxation rate of the Gd-doped sample is larger than that of the pure sample

The muon spin precesses around a local magnetic field, B_μ (with a frequency $\nu = (\gamma_\mu/2\pi)|B_\mu|$, where $\gamma_\mu/2\pi = 135.5\,\text{MHz}\,\text{T}^{-1}$). This local field ($B_\mu \approx 0.22\,$T at $T=0$) is a sum of various terms, including the Lorentz field $B_{\rm L}$, the hyperfine field $B_{\rm hf}$, the demagnetizing field $B_{\rm demag}$ and the dipolar field $\mathbf{B}_{\rm dip}$. The latter quantity is a function of the muon-site $\mathbf{r}_\mu$ and can be written as

$$B^{\alpha}_{\rm dip}(\mathbf{r}_\mu) = \sum_i D_i^{\alpha\beta}(\mathbf{r}_\mu)\, m_i^{\beta}, \tag{1}$$

a sum over the magnetic ions in the crystal; the magnetic moment of the ith ion is $\mathbf{m}_i$. In Eq. (1), $D_i^{\alpha\beta}(\mathbf{r}_\mu)$ is the dipolar tensor given by $D_i^{\alpha\beta}(\mathbf{r}_\mu) = \frac{\mu_0}{4\pi R_i^3}\left(\frac{3R_i^\alpha R_i^\beta}{R_i^2} - \delta^{\alpha\beta}\right)$, where $\mathbf{R}_i \equiv (R_i^x, R_i^y, R_i^z) = \mathbf{r}_\mu - \mathbf{r}_i$. The behaviour of this tensor is dominated by the arrangement of the nearest-neighbour magnetic ions and leads to a non-zero local magnetic field for almost all possible muon sites [20]. On electrostatic grounds, the likely muon site in EuO is at the $\frac{1}{4}\frac{1}{4}\frac{1}{4}$ position [see Fig. 2(a)], equidistant from four Eu cations and four oxygen anions. Because of the magnetic anisotropy [21], the easy axis for the Eu moments is along the $\langle 111\rangle$ set of directions and for this moment alignment the $\frac{1}{4}\frac{1}{4}\frac{1}{4}$ position is a point at which the dipolar magnetic field actually vanishes. The value of $B_{\rm dip}$ has been calculated for the case in which the muon is displaced from the $\frac{1}{4}\frac{1}{4}\frac{1}{4}$ site and its position is allowed to vary along the [111] direction, see Fig. 2(b). The dipolar field vanishes at both $\frac{1}{4}\frac{1}{4}\frac{1}{4}$ (muon site) and $\frac{1}{2}\frac{1}{2}\frac{1}{2}$ (oxygen site) and increases sharply as the site moves away from these special positions of high symmetry. The two curves show the cases in which the muon displacement is the same or a different choice of $\langle 111\rangle$ direction. In the former case, the dipolar field at the muon site is parallel to the moment direction; in the latter case (for which there are three possibilities), it lies along one of the crystallographic axes and its amplitude is reduced by a factor of $\sqrt{3}$. Thus if the muon site is displaced from the $\frac{1}{4}\frac{1}{4}\frac{1}{4}$ position towards a particular oxygen anion, then there would be a contribution to the dipole field resulting in two precession frequencies with a ratio of $\sqrt{3}$, and with amplitudes in a 3:1 ratio. Since only a single frequency is observed, we conclude that the muon site is indeed at the

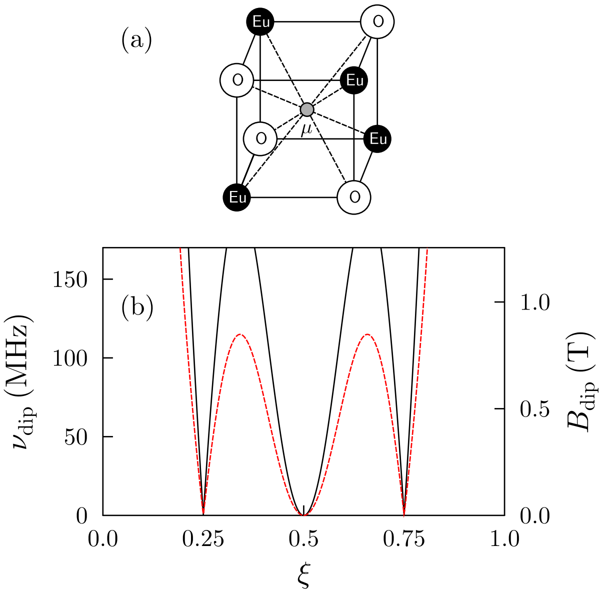

FIG. 2: (Color online.) (a) Muon site in EuO. The crystal structure for one-eighth of the unit cell is shown (the side of the cube is $a/2$). (b) Calculated dipole field along the $\langle 111\rangle$ axes in EuO. The muon is located at $\xi\xi\xi$ and the Eu moments are all aligned along [111] (solid black line) or one of [$\bar{1}$11], [1$\bar{1}$1] or [11$\bar{1}$] (dashed red line).

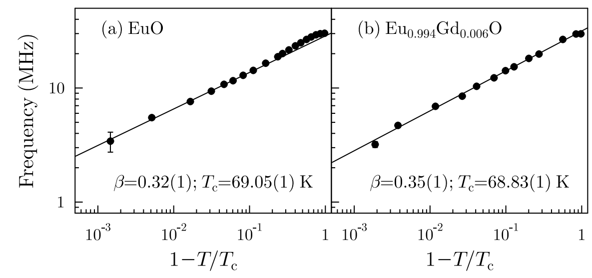

FIG. 3: Precession frequency extracted from μSR data as a function of temperature close to $T_{\rm C}$ for (a) EuO and (b) $\mathrm{Eu_{0.994}Gd_{0.006}O}$.

$\frac{1}{4}\frac{1}{4}\frac{1}{4}$ position in which the dipolar field is zero, so that the observed local field ($B_\mu = 0.22\,\mathrm{T}$ at $T = 0$) is due to a sum of the Lorentz field ($B_{\rm L} = \mu_0 M/3 = 0.80\,\mathrm{T}$ at $T = 0$), $B_{\rm demag}$ and $B_{\rm hf}$. Since the sample is polycrystalline and multidomain, we neglect $B_{\rm demag}$ and deduce that the hyperfine field $B_{\rm hf} < 0$ (antiparallel to the magnetization), as found for EuS [22], and takes the value $B_{\rm hf} = -B_{\rm L} \pm B_\mu$, and so either -0.58 or $-1.02\,\mathrm{T}$. For both samples, the amplitude of the oscillatory component is reduced from the full value at low temperature [see Fig. 1(a,b)], but recovers on warming towards $T_{\rm C}$, so a fraction of muons may implant in some additional state which depolarizes the muon very rapidly.

We note that a recent experiment [23] on SmS has shown evidence for the formation of a bound magnetic polaron consisting of an electron around the implanted muon, in which the electron localization is stabilised by exchange energy. This occurs in the paramagnetic state in which a ferromagnetic droplet is localized in the paramagnetic host. A similar effect has been noted in EuS [24] although the larger magnetization ensures it occurs at temperatures $\gg T_{\rm C}$ [25] and the same will be true in EuO in which the magnetization is even larger. Therefore such a muon-related polaron is not relevant for EuO in the studied temperature regime.

The temperature dependence of the precession frequency for both EuO and $\mathrm{Eu_{0.994}Gd_{0.006}O}$ was followed near $T_{\rm C}$ and the results are plotted in Fig. 3. The fitted values of $T_{\rm C}$ and the critical exponent β are similar in each case, though the value of β is quite sensitive to the precise value taken for $T_{\rm c}$. Due to the difficulty in stabilising the temperature better than $\approx 10\,\mathrm{mK}$, we do not believe that the difference between the two values of β is significant. They are both close to 0.36–0.37 obtained using neutron scattering [26] and 0.38 obtained from a second order ϵ expansion for the Heisenberg ferromagnet with dipolar interactions [27].

Above $T_{\rm C}$, we observe simple exponential relaxation [Fig. 1(a,b)] with a relaxation rate λ. For zero-field relaxation of muons initially polarized parallel to z, λ can be written in terms of field-field correlation functions using $\lambda = \frac{\gamma_\mu^2}{2}\int_{-\infty}^{\infty} \mathrm{d}t\, \left(\langle B_x(0)B_x(t)\rangle + \langle B_y(0)B_y(t)\rangle\right)$ [18]. When each Eu spin component fluctuates, it produces a field fluctuation via the resulting modulation of the dipolar and hyperfine couplings. Our measurements of the zero-field relaxation rate for the EuO sample are plotted in Fig. 4 (a less complete set of data for the Gd-doped sample is also shown). There is a small rise in λ as the temperature is lowered towards $T_{\rm C}$ but apart from this λ remains just below $\approx 2\,\mathrm{MHz}$ for both samples across the entire range studied.

These results can be compared with calculations on a localized Heisenberg ferromagnet which have been performed with EuO in mind [15, 16] (Fig. 4). The theory of Lovesey and Engdahl [15] includes only the dipolar coupling and has been evaluated for temperatures above $1.3T_{\rm C}$, assuming a muon site of $\frac{1}{4}\frac{1}{4}\frac{1}{4}$. Though underestimating the observed experimental values, this theory does remarkably well in providing a good estimate of the rough size of λ, the discrepancy perhaps being due to neglecting the hyperfine contribution. It is known that critical fluctuations enhance the role of the hyperfine coupling over the dipole coupling [14] because $B_{\rm dip} = 0$ at the muon site in the ordered state and the peak in the susceptibility is at $\mathbf{k} = 0$. Nevertheless, when the dipolar calculation is extended into the critical regime (just above, and very close to, $T_{\rm C}$) it predicts a divergence in λ which is not observed. An earlier mode-coupling approach [16] also predicts a very sharp increase in λ on

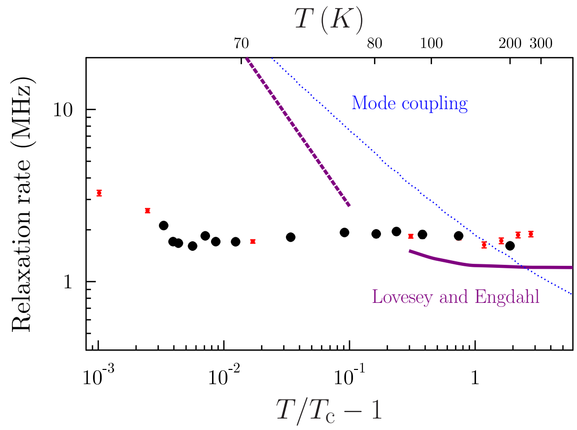

FIG. 4: (Color online.) Relaxation rate as a function of temperature in the paramagnetic regime for pure EuO (solid circles) and (b) $Eu_{0.994}Gd_{0.006}O$ (dots). The predicted relaxation rates for $T > T_C$ according to Ref. 15 (numerical calculation: thick solid line, purple; critical regime using experimental values of correlation length: thick dashed line, purple) and Ref. 16 (dotted line, blue) are also shown.

cooling to T_C from about 0.3 K above it; this is also not observed in our data. Magnetic polaron formation has been detected using Raman scattering [28] in a narrow range ($\approx$ 20 K) above T_C. It may be that the formation of magnetic polarons modifies the relaxation in this regime from that which would be expected from theory, perhaps by providing an additional relaxation channel for the muon which masks the critical slowing down predicted by the theory and hence the absence of the divergence in λ. We note that a similar absence of a divergence in λ is observed in EuB_6 [29] in which magnetic polarons have been found [28].

In conclusion, we have identified the muon site in EuO and estimated the hyperfine field. Our results confirm long-range order which is relatively insensitive to low doping of Gd. The measured λ in the paramagnetic state agrees quite well with the theory of Lovesey and Engdahl, but the available theories fail in the critical regime, possibly due to magnetic polaron formation.

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